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Simulating present-day and future air quality as climate changes: Model evaluation

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Abstract

The global-regional climate-air pollution modeling system (GRE-CAPS) has been developed, coupling an existing general circulation model/chemical transport model (GCM/CTM), a regional meteorological model, and a regional chemical transport model. This system is intended to enable studies of the effects of changes in climate, intercontinental transport, and emissions on regional and urban air quality. The GRE-CAPS system consists of the GISS II′ GCM/CTM, the MM5 regional meteorological model, and the PMCAMx regional CTM. The modeling system is evaluated for the present day, with comparisons between model-predicted, measured ozone, and speciated $PM_{2.5}$ concentrations. The ability of the model to predict present-day concentrations of ozone and $PM_{2.5}$ is compared to that of PMCAMx when used for retrospective modeling. Comparisons between model-predicted temperatures and precipitation are also made. The model was used to simulate five present-day Januaries and six present-day Julys. The biases and errors in GRE-CAPS-predicted ozone concentrations were similar to those of PMCAMx when used for standard retrospective modeling. The fractional biases in mean daily peak ozone concentration and mean daily maximum 8-h average ozone concentration are both $<10\%$. The model-predicted distribution of peak hourly and daily maximum 8-h average values agreed rather well with the measured distribution. There is less agreement between the model and measurements in the number of hours with ozone mixing ratios >70 or 80 ppb, though this is also the case with standard PMCAMx modeling. The predictions of $PM_{2.5}$ concentrations by GRE-CAPS were also of similar quality to those of PMCAMx driven by historical meteorology. The fractional biases in the predictions of total $PM_{2.5}$, sulfate, ammonium, and nitrate were all $<25\%$ in both January and July. The model agrees well with organic $PM_{2.5}$ measurements from the IMPROVE network, though there is less agreement with measurements from the STN network. The GRE-CAPS system is shown to reproduce ozone and $PM_{2.5}$ concentrations for the present day rather well, with model performance similar to that of PMCAMx for standard retrospective episode modeling with historical meteorology. GRE-CAPS will be used in future studies to examine the effects of changes in climate, global emissions, and intercontinental transport on regional air quality.

Keywords: Climate change; Ozone; Particulate matter; Model evaluation

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1. Introduction

Much recent work has focused on the effect that changes in meteorology and climate can have on ozone (O_3) and particulate matter (PM) concentrations. Observational statistical studies (Bloomfield et al., 1996; Guicherit and van Dop, 1977; Wise and Comrie, 2005) have examined the links between meteorology and O_3 or PM concentrations and found that air quality can change significantly with changes in meteorology. The key links between meteorology and O_3 and PM concentrations over the Eastern USA were investigated in a series of sensitivity studies with the chemical transport model (CTM) PMCAMx by Dawson et al. (2007a, b). Temperature had the major meteorological effect on O_3 concentrations with an increase in maximum daily 8-h average (MDA8) O_3 of $0.34\,ppb\,K^{-1}$ (Dawson et al., 2007a, b), while several meteorological parameters had appreciable effects on $PM_{2.5}$ concentrations, depending on the season and the relative importance of the various PM species (Dawson et al., 2007a, b). Changes in temperature, precipitation, wind speed, and mixing height were all shown to have potentially significant impacts on air quality in these studies.

Recent research has also focused on the effects of global climate and transport on regional air quality. The effect of intercontinental transport of O_3 and precursors has been the subject of recent work (Jacob et al., 1999; Fiore et al., 2002, 2003). These authors have shown that intercontinental transport can affect O_3 concentrations in the USA and that changes in European and Asian emissions can increase O_3 in the USA by several ppb. Prather et al. (2003) examined the effects of global future emission changes, using the IPCC (2000) SRES A2 emissions scenario, on O_3 concentrations globally and calculated a potentially large increase in O_3 concentrations.

In a global modeling study with changing climate but constant anthropogenic emissions, Racherla and Adams (2006) identified changes in water vapor as a major explanation for future decreases in global tropospheric O_3 burdens in the 2050s compared to the present day. The same study, however, also predicted increase in average O_3 concentrations of several ppb over the Eastern US, with the largest increase during the summer. Racherla and Adams (2006) also linked changes in fine PM concentrations largely to changes in precipitation. Global burdens in the 2050s of all PM species were predicted to decrease by 2–18% due to increased wet deposition resulting from increased global average precipitation and changes in aerosol volatility due to temperature changes. The effects of changes in climate on O_3 were studied by Murazaki and Hess (2006), who reported that temperature, water vapor, cloud cover, transport, and lightning NO_x changes are having significant effects on O_3. The same work also calculated, using the IPCC (2000) A1 scenario, that background O_3 over the USA would decrease over the next century by 0–2 ppb, but that O_3 produced in the USA would increase up to 6 ppb.

Hogrefe et al. (2004a, b) used meteorology generated by the GISS GCM (Russell et al., 1995) and downscaled by a regional meteorological model (MM5) (Grell et al., 1994) to simulate present-day and future climates in a regional CTM (CMAQ) (Byun and Ching, 1999). The purpose of this work was to simulate present-day and future O_3 concentrations over the eastern USA, using the IPCC (2000) A2 scenario for the future up to the 2080s. This modeling system reasonably reproduced present-day summertime meteorology and O_3 concentrations (Hogrefe et al., 2004a, b) and predicted an increase in summertime MDA8 O_3 in the 2050s compared to the present day of 4.2 ppb (Hogrefe et al., 2004a, b). This work concluded that changes in climate alone could have an appreciable effect on future O_3 concentrations.

Tagaris et al. (2007) also used downscaled meteorology to simulate O_3 and $PM_{2.5}$ in the present day and in 2050, using the IPCC A1B scenario. This coupled modeling system overpredicted summertime MDA8 O_3 by an average of 15% and underpredicted summertime $PM_{2.5}$ by an average of 30%. The same study also predicted a rather small impact of climate on pollutant concentrations, with predicted emission changes having a much larger impact. The effects of climate alone on air quality included a very small change in O_3 concentrations in the summer and decreases in summer $PM_{2.5}$ due to increased precipitation.

Coupled modeling systems use GCM-predicted meteorology that is not year-specific, in contrast to traditional retrospective modeling in which the CTM uses assimilated meteorology. This type of coupled modeling system has been shown to simulate O_3 concentrations accurately, but no such studies have been performed for PM. A coupled model with a GCM/CTM and a regional CTM that includes aerosol species will allow the effects of

changes in climate and intercontinental transport on both O_3 and PM concentrations to be studied.

The primary goal of this work is to develop and evaluate a coupled global–regional modeling system, joining previously evaluated models, to simulate present day and future concentrations of O_3 and $PM_{2.5}$. Concentrations predicted for the present day by GRE-CAPS were compared to measured concentrations in the eastern USA. Multiple years were simulated so that both mean concentrations and the interannual variability in concentrations could be evaluated. The ability of GRE-CAPS, which uses GCM-predicted meteorology to drive a regional CTM, to capture present-day O_3 and $PM_{2.5}$ concentrations is compared to the ability of a regional CTM, which uses historical meteorology, to capture episodic conditions correctly. Also, long-term climatological average concentrations are considered, not just short-term episodic concentrations. GRE-CAPS will be used in future work to examine the effects of climate change as well as changes in emissions and intercontinental transport on regional air quality. In order to compare other scenarios to the present day in future studies, the ability of GRE-CAPS to simulate the present day accurately needs to be evaluated.

2. The GRE-CAPS modeling system

The GRE-CAPS modeling system is comprised of three models (Fig. 1), spanning the global to the regional scale. The GISS II′ GCM/CTM is used to generate the climate and pollutant concentration fields at the global scale. Details of the global model utilized in the current study are provided by Racherla and Adams (2006). The global model (Liao et al., 2003, 2004) consists of: (1) the Goddard Institute for Space Studies general circulation model II′ (GISS GCM II′) (Hansen et al., 1983; Rind and Lerner, 1996; Rind et al., 1999); (2) the Harvard tropospheric O_3–NO_x–hydrocarbon chemical model (Mickley et al., 1999); and (3) an aerosol model including sulfate, nitrate, ammonium, black carbon, and organic carbon (Adams et al., 1999; Chung and Seinfeld, 2002; Liao et al., 2003, 2004).

The version of GISS GCM II′ incorporated in the current study is an atmosphere-only GCM, and it utilizes specified monthly-mean ocean boundary conditions. It has a horizontal resolution of 4° latitude by 5° longitude, with nine vertical layers centered at 959, 894, 786, 634, 468, 321, 201, 103, and 26 hPa. Necessary GCM variables are passed to the tropospheric gas-phase chemistry and aerosol modules every 4 h. The model transports 88 species; of these, 24 species are used to describe O_3–NO_x–hydrocarbon chemistry, and the remaining is for the simulation of the aerosols, including sulfate, nitrate, ammonium, black carbon, primary and secondary organic aerosol, sea salt, and dust.

The global-scale anthropogenic emissions used in the model, which correspond to present day, are summarized in Liao et al. (2003, 2004). Climate-sensitive emissions include isoprene (Guenther et al., 1995; Wang et al., 1998), lightning and soil NO_x (Wang et al., 1998), DMS (Kettle et al., 1999), sea salt and mineral dust (Liao et al., 2004). The dry deposition of all gas-phase species is determined based on the resistance-in-series scheme of Wesely (1989), and the wet deposition is coupled with the

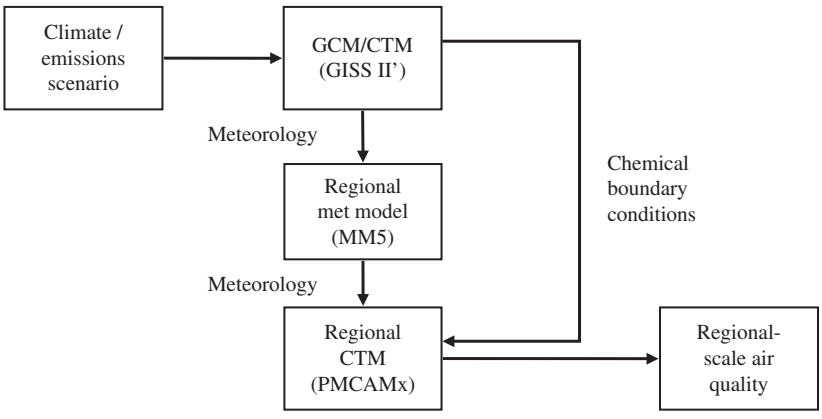

Fig. 1. Schematic representation of GRE-CAPS system.

GCM treatment of clouds and precipitation (Koch et al., 1999; Del Genio and Yao, 1993; Del Genio et al., 1996).

The global-scale meteorology is downscaled to the regional scale by using the MM5 meteorological model. Meteorological conditions on the outer edge of the entire USA modeling domain were used as boundary conditions for the MM5 simulations. MM5 simulations used a 108-km-resolution grid over the entire USA and a 36-km-resolution nested grid over the Eastern USA. The MM5 configuration included the Eta planetary boundary layer scheme (Janjic, 1990, 1994) and the rapid radiative transfer model (RRTM) for short-wave and long-wave radiation transfer (Mlawer et al., 1997). The MM5 model also used the Kain–Fritsch cumulus parameterization (Kain and Fritsch, 1994) and the Noah land-surface model (Chen and Dudhia, 2001a, b). The GCM and MM5 grid alignments were reconciled, and the coarse GCM-predicted meteorology was interpolated to match the relatively fine MM5 vertical scale, using standardized utilities available from the University Corporation for Atmospheric Research (UCAR). The REGRID program (UCAR, 2004a, b) was used to read the GCM-predicted meteorology and to interpolate those predictions onto the horizontal MM5 grid and map projection. The INTERPF program (UCAR, 2004a, b) performed the vertical interpolation, diagnostic computation, and data formatting necessary to create the initial conditions and lateral and lower boundary conditions for the MM5 simulation.

The regional CTM used in GRE-CAPS is PMCAMx. The development and evaluation of the PMCAMx model have been described by Gaydos et al. (2007) and Karydis et al. (2007). PMCAMx uses the framework of CAMx v. 4.02 (Environ International Corporation, 2004) to simulate horizontal and vertical advection, horizontal and vertical dispersion, wet and dry deposition, and gas-phase chemistry. The Carbon-Bond IV (CB4) mechanism (Gery et al., 1989) with updated isoprene chemistry (Carter, 1996), including 34 gas-phase and 12 radical species, is used for gas-phase chemistry calculations. The aerosol processes included are summarized in Gaydos et al. (2007), with nitrate chemistry improvements by Karydis et al. (2007). The model uses ten aerosol size sections, spanning diameters from 40 nm to 40 μm, six of which comprise $PM_{2.5}$. Inorganic aerosol formation was simulated using the bulk equilibrium approach of Capaldo et al. (2000), while aqueous chemistry was modeled using the variable size resolution model (VSRM) of Fahey and Pandis (2001). Equilibrium between the gas and aerosol phases for organics was calculated using the secondary organic aerosol model (SOAM II) of Strader et al. (1999) and implemented by Koo et al. (2003). Primary organic aerosol (POA) was treated as nonvolatile, while secondary organic aerosol (SOA) was treated as semi-volatile.

The PMCAMx modeling domain was the Eastern USA (Fig. 2) with a $36 \times 36\,km^2$ resolution grid. Fourteen vertical layers were used, up to an altitude of approximately 6 km. The emissions inventory used was the Midwest Regional Planning Organization's Base E inventory (LADCO, 2003), including BIOME3 biogenics (Wilkinson and Janssen, 2001). All emissions, including biogenics, were for July 2001 or January 2002. The same emissions were used for all simulations of the present, regardless of meteorology.

Output from both the global GCM/CTM and the regional meteorological model were used as input into the regional CTM. The global-model-predicted chemical concentrations around the outside of the Eastern USA modeling domain were used as boundary conditions for the PMCAMx simulations. Generally, individual organic species from the GCM/CTM were lumped into PMCAMx CB4 species (Table 1). Concentrations of aerosol species, most of which were treated in the global model as bulk PM and some of which (sea salt and dust) were treated using size sections, were split into the PMCAMx size sections (Tables 2 and 3). The bottom five vertical layers of the GCM/CTM were split among the fourteen PMCAMx layers, without interpolation or smoothing. The eastern and western boundaries of the PMCAMx domain were the length of eight GCM/CTM cells, while the northern and southern boundaries of the PMCAMx domain were the width of ten GCM/CTM cells. Chemical (gas and aerosol) boundary conditions were updated every 4 h. Due to an overprediction of wintertime dust in the GCM/CTM, January dust concentrations were reduced by a factor of 10 before being used as PMCAMx boundary conditions. The downscaled hourly meteorology from MM5 was used as the meteorological input to PMCAMx. Six Julys and five Januaries were simulated in PMCAMx. Each July simulation was 25 days long, and each January simulation was 26 days long (plus 3 days of model spin-up).

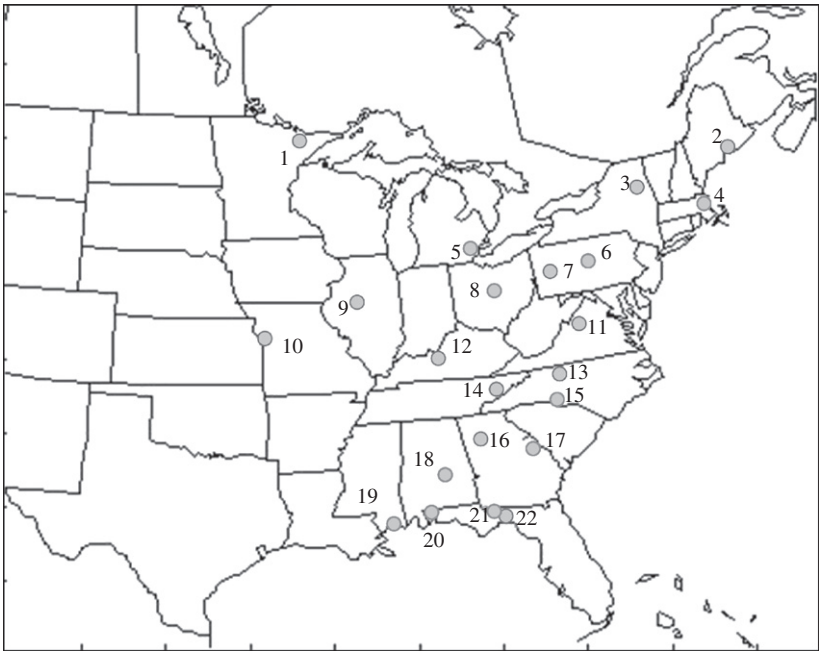

Fig. 2. Map of Eastern USA modeling domain and locations of 22 measurement sites. "I" denotes an IMPROVE site and "S" denotes an STN site. 1. Boundary Waters Area, Minnesota (I); 2. Acadia NP, Maine (I); 3. Whiteface Mountain, New York (S); 4. Boston, Massachusetts (S); 5. Detroit, Michigan (S); 6. State College, Pennsylvania (S); 7. Pittsburgh, Pennsylvania (S); 8. Columbus, Ohio (S); 9. Decatur, Illinois (S); 10. Kansas City, Missouri (S); 11. Shenandoah NP, Virginia (I); 12. Mammoth Cave NP, Kentucky (I); 13. Winston-Salem, North Carolina (S); 14. Great Smoky Mountains NP, Tennessee (I); 15. Charlotte, North Carolina (S); 16. Atlanta, Georgia (S); 17. Augusta, Georgia (S); 18. Montgomery, Alabama (S); 19. Biloxi, Mississippi (S); 20. Pensacola, Florida (S); 21. Tallahassee, Florida (S); 22. St. Marks, Florida (I).

Table 1
Grouping of gas-phase species in GISS-II′ GCM and PMCAMx

GISS species	PMCAMx species	Explanation
NO	NO	
NO_2	NO_2	
O_3	O_3	
PAN, PPN	PAN	Peroxyacylnitrates
CO	CO	
ALK_4, C_3H_8, C_2H_6	PAR	Parrafins
PRPE	OLE	Olefins
ISOP, HC I–V	OLE_2	Biogenics
CH_2O	FORM	Formaldehyde
CH_3CHO	ALD_2	Higher aldehydes
HNO_3	HNO_3	
PMN, R_4N_2	NTR	Organic nitrates
H_2O_2	H_2O_2	
SO_2	SO_2	
NH_3	NH_3	

Table 2
Grouping of aerosol species in GISS II′ GCM and PMCAMx

GISS	PMCAMx
SOA	Split equally among 4 PMCAMx SOA species. Also, split into the six fine size bins. Fractions of mass into each bin: 0.1, 0.1, 0.1, 0.2, 0.3, 0.2
POC, PEC	Split into the six fine size bins. Fractions of mass into each bin: 0.1, 0.1, 0.1, 0.2, 0.3, 0.2
Dust	Sectional, grouped with PMCAMx species CRST
Sea salt	Sectional, split into Na and Cl
Ammonium, non-dust nitrate, non-dust non-sea-salt sulfate	Split into ten size bins: 0.1, 0.1, 0.1, 0.2, 0.25, 0.15, 0.05, 0.02, 0.02, 0.01
Dust and sea-salt sulfate, dust nitrate	Split equally among five largest size bins
Alkaline calcium	Split equally among four largest size bins, grouped into CRST

3. Measurements

The 22 locations examined were spread throughout the domain and consisted of a mix of urban, rural, and remote locations (Fig. 2). The monitors at

Table 3
PMCAMx aerosol size sections

Section	Diameter range
1	40–80 nm
2	80–160 nm
3	160–320 nm
4	320–625 nm
5	625 nm–1.25 μm
6	1.25–2.5 μm
7	2.5–5 μm
8	5–10 μm
9	10–20 μm
10	20–40 μm

these locations measured 24-h average concentrations of $PM_{2.5}$ species including sulfate, nitrate, ammonium, organic carbon (OC), and total $PM_{2.5}$ approximately every 3 days on average. The PM measurements were from EPA speciation trends network (STN) monitors from 2002 to 2005 or interagency monitoring of protected visual environments (IMPROVE) monitors from 2001 to 2004. All 22 locations also had a nearby or co-located O_3 monitor. Ammonium concentrations were not measured at the Boundary Waters, Acadia, or St. Marks sites. Of the 22 sites, 16 were from the STN and six were from the IMPROVE network. The 19 sites with ammonium measurements were selected because they were the only sites with several years of speciated PM measurements (including ammonium) and a co-located O_3 monitor. The sites without ammonium measurements were added to allow a greater area of the domain to be studied.

The IMPROVE measurements for organic carbon (OC) were blank corrected and multiplied by 1.4 to yield estimates of OM. STN OC measurements were corrected using an average value of $0.9\,\mu g\,C\,m^{-3}$ that was the average blank value measured for the 16 STN sites. For consistency, STN OC measurements were multiplied by the same factor of 1.4 to estimate OM. Because of the differences in blank correction and collection methods, IMPROVE and STN organic measurements were analyzed separately in the model evaluation.

Only PM concentrations were compared in January, while both O_3 and PM concentrations were compared in July. The peak hourly O_3 concentration, MDA8 O_3, hours with O_3 mixing ratio >70 ppb, and hours with O_3 mixing ratio >80 ppb were used in the model evaluation; monthly average $PM_{2.5}$ concentrations and the distribution of 24-h average $PM_{2.5}$ concentrations were used in the analysis due to the long time between measurements. Both the means and interannual variabilities of the various model-predicted metrics were compared to those of the measured values so that the ability of GRE-CAPS to capture both climatological averages and interannual variabilities could be evaluated.

4. Model evaluation

A 10-year simulation of the present day was run using the GCM/CTM. The ocean boundary conditions for this simulation were obtained from a transient simulation performed using a fully coupled atmosphere–ocean GCM (the GISS Model III [Russell et al., 1995; R. Healy, personal communication, 2005]), as described by Racherla and Adams (2006). The ocean boundary conditions in the present study correspond to a decadal average of the 1990s and 2050s from the above simulation, respectively, with month-to-month variability. The meteorology predicted by the GCM/CTM was downscaled to the Eastern USA modeling domain with MM5. The first MM5 year was disregarded as spinup, and the next five Januaries and six Julys were passed on to PMCAMx for simulation. The predicted meteorology, O_3 concentrations, and $PM_{2.5}$ concentrations for the simulated months were compared to measurements. The mean error (ERROR), mean bias (BIAS), fractional error (FERROR), and fractional bias (FBIAS) were calculated to assess the model performance:

$$\mathrm{ERROR} = \frac{1}{N}\sum_{i=1}^{N}|P_i - O_i|, \quad \mathrm{BIAS} = \frac{1}{N}\sum_{i=1}^{N}(P_i - O_i)$$

$$\mathrm{FERROR} = \frac{2}{N}\sum_{i=1}^{N}\frac{|P_i - O_i|}{P_i + O_i}, \quad \mathrm{FBIAS} = \frac{2}{N}\sum_{i=1}^{N}\frac{P_i - O_i}{P_i + O_i}$$

where N is the total number of observations, P_i is the model-predicted value of the quantity, and O_i is the corresponding observed value of the quantity.

4.1. Meteorology

The meteorology generated by downscaling the GCM predictions was compared to observed meteorology. The comparison of 5 years of National Climate Data Center observational data (2001–2005) from Kansas City, Atlanta, Boston,

and Pittsburgh to model predictions in the same sites is discussed here for illustrative purposes. The variables examined were mean daily minimum temperature, daily mean temperature, mean daily maximum temperature, and total monthly precipitation. The temperature comparisons for January and July are shown in Table 4. The temperature data show a cold bias in the model-predicted temperatures that was stronger in July than in January. The interannual variabilities of the temperature data were also compared; the variances of the sets of model-predicted monthly average daily minimum and maximum temperatures were compared to the variances of the same measured temperatures using *F*-tests at the 5% significance level. The variability of January minimum temperatures was significantly overpredicted in Kansas City only, while the variability of January maximum temperatures was significantly overpredicted in Kansas City and Atlanta. The interannual variabilities of both average maximum and minimum temperatures were significantly overpredicted for all four cities in July. Most of the large interannual variability originated in the regional meteorological model, not in the GCM. For these four locations, the bias in average January temperature was −1.0 °C, while the bias in average July temperatures was −2.7 °C. These biases are of similar magnitude to those of other modeling systems, such as those of Leung et al. (2004) and Leung and Gustafson (2005).

The model-predicted and measured amounts of precipitation were also compared. Average model-predicted and measured total precipitation and the range of monthly precipitation totals for January and July are shown in Fig. 3. Model-predicted precipitation totals do not include snow since snow is not included in the PMCAMx model. The predicted precipitation was underestimated in all four cities in January, indicating a dry wintertime bias, though part of this bias is likely due to the lack of snow. In July, total precipitation was underestimated in Atlanta and Kansas City and overestimated in Pittsburgh and Boston. These differences indicate large biases in specific locations, but smaller average biases across the four locations: −41% in January and +13% in July. Large biases in precipitation are common for similar coupled modeling systems. Leung and Gustafson (2005) calculated dry biases of 50–80% over parts of the Eastern USA during summer, and Leung et al. (2004) calculated wet biases in parts of the Northwestern USA during winter of up to 50%. These biases in meteorological model prediction can cause significant errors in the PM concentrations predicted by the regional chemical transport model, even in retrospective modeling studies, such as in Gaydos et al. (2007).

4.2. Ozone

In general, GRE-CAPS performed nearly as well as standard PMCAMx in simulating O_3 concentrations. The full set of measured and model-predicted daily maximum hourly and 8-h average O_3 concentrations for the 22 locations were compared

Table 4
Model-predicted and measured mean daily minimum and maximum temperatures (°C) for five Januaries and five Julys

	January			July		
	Mean min *T*	Mean *T*	Mean max *T*	Mean min *T*	Mean *T*	Mean max *T*
Atlanta						
Model	0.9	4.8	8.7	18.6	23.5	28.4
Measured	1.1	7.5	11.8	21.6	26.2	30.8
Kansas City						
Model	−6.4	−2.4	1.5	19.5	25.4	31.3
Measured	−6.5	−1.5	3.4	20.6	26.3	32.0
Pittsburgh						
Model	−5.0	−2.1	0.7	15.0	19.0	23.1
Measured	−6.4	−2.5	1.3	17.3	22.7	28.0
Boston						
Model	−7.8	−3.4	−0.9	16.0	19.2	22.3
Measured	−6.0	−2.4	1.1	18.3	22.6	27.0

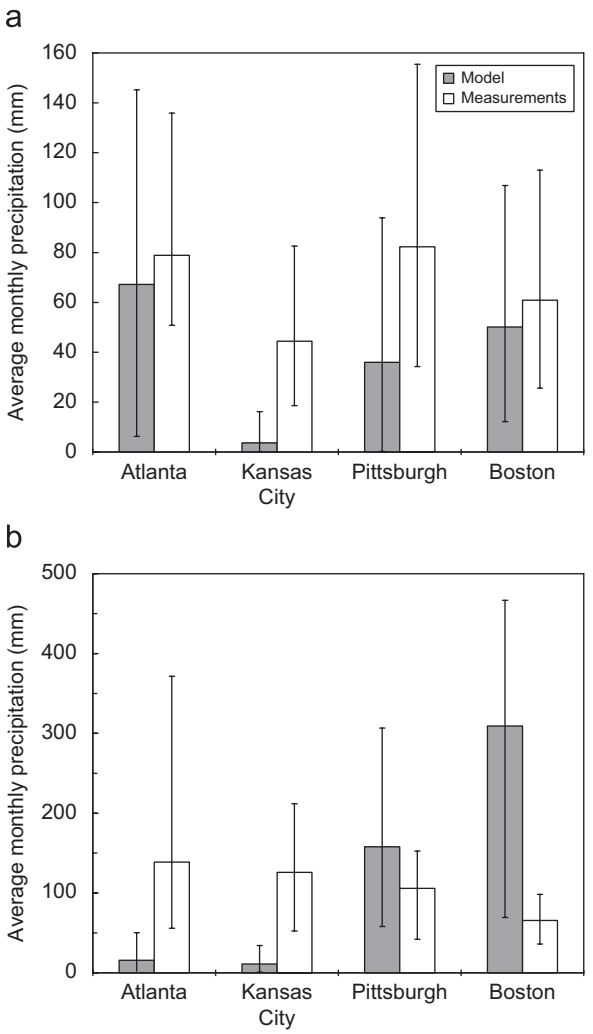

Fig. 3. Average model-predicted and measured monthly precipitation for (a) January and (b) July. Error bars indicate full range of monthly precipitation.

using cumulative distribution functions (CDFs). The CDFs for all six model-predicted Julys and all five measured Julys are shown in Fig. 4. The larger spread among the model-predicted years compared to the measured years indicates an overprediction of the interannual variability of O_3 concentrations. Much of the overprediction in O_3 variability is due to the single July farthest to the right in Fig. 4. This variability can be attributed in large part to the interannual variability in meteorology. The total flux of O_3 and precursors entering from the boundaries of the domain varied by $<20\%$ from simulation to simulation.

The biases and errors for the four O_3 metrics are shown in Table 5. The 1- and 8-h metrics were predicted rather well, while the hours over 70 or 80 ppb were predicted less accurately. All four metrics had a positive bias, indicating that overpredictions of O_3 concentrations were dominant. The biases and errors in the 1- and 8-h metrics were similar to those seen by Hogrefe et al. (2004a, b). When the total sets of modeled and measured mean MDA8 O_3, mean peak O_3, number of hours >80 ppb, and number of hours >70 ppb for each July were compared using *F*-tests, the variances of

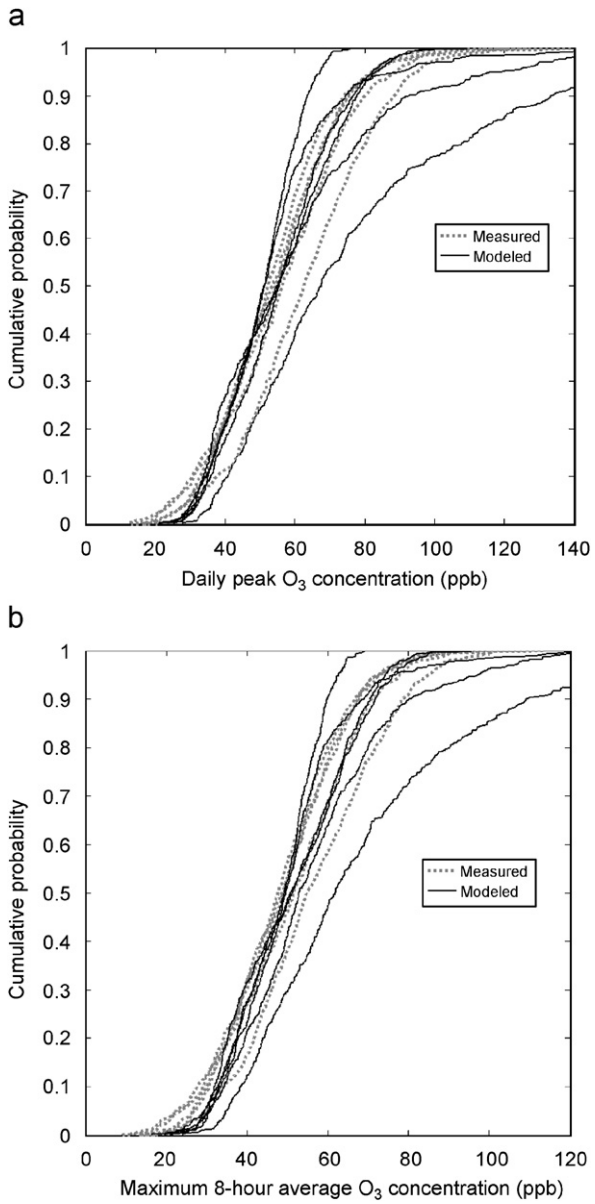

Fig. 4. Cumulative distribution functions of daily peak (a) hourly and (b) 8-h O_3 concentrations for six model-predicted and five measured Julys.

the model predictions were significantly greater ($p<0.05$) than the variances of the measurements. Excluding the one especially high-ozone year, the fractional biases in the peak, MDA8, and hours >70 ppb metrics were all $<3\%$, though the bias in the hours >80 ppb and the fractional errors for all four metrics remained essentially unchanged.

The model-predicted average daily peak and MDA8 O_3 concentrations are compared to measured values in Fig. 5. The large majority of model-predicted average concentrations are within 30% of the measured values. The closest agreement between model and measurements was in the area from New York, Pennsylvania, and Virginia west to Illinois. The largest overpredictions were at the Southeastern sites. Mean daily peak and MDA8 concentrations were overpredicted in 13 locations and underpredicted in 9. The mean number of hours >70 ppb was overpredicted in 12 locations and underpredicted in 10, while the mean number of hours >80 ppb was overpredicted in 15 locations and underpredicted in 7. The differences were statistically significant (using a t-test with $p<0.05$) in at most four locations for each of the O_3 metrics, however.

The model significantly overpredicted the inter-annual variability in many locations. The variance of the set of model-predicted monthly average daily peak O_3 was significantly overpredicted in 11 locations, while the variance of the model-predicted MDA8 concentrations was significantly overpredicted in 10 locations. The variance of the monthly number of hours over 70 ppb was significantly overpredicted in 8 locations and underpredicted in 2, while the variance in the number of hours over 80 ppb was significantly overpredicted in 11 locations and underpredicted in 4.

The discrepancies between modeled and measured O_3 concentrations are likely due to impacts from both the regional CTM itself and the down-scaled meteorology. In traditional retrospective simulations of July 2001, Dawson et al. (2007a, b) and Gaydos et al. (2007) were generally able to capture O_3 concentrations in Atlanta, Pittsburgh, and Kansas City, though these simulations also had difficulty in producing the number of hours with O_3 concentrations >70 or 80 ppb. In modeling the number of hours with $O_3>70$ or 80 ppb in Atlanta, Pittsburgh, and Kansas City, PMCAMx had large errors similar to those of the GRE-CAPS system. The biases of the GRE-CAPS model are of similar magnitude to those of PMCAMx alone. The discrepancies between historical and model-predicted meteorology, especially temperature, contribute additional errors in O_3 concentration predictions but do not dominate the overall modeling error. Part of the discrepancy may also be due to the limitations of the CB4 mechanism.

The ability of GRE-CAPS to capture O_3 concentrations was evaluated using the performance criteria based on the model fractional error and bias introduced by Morris et al. (2005). The performance of GRE-CAPS is summarized in Table 6. The 1- and 8-h concentrations of O_3 were captured quite

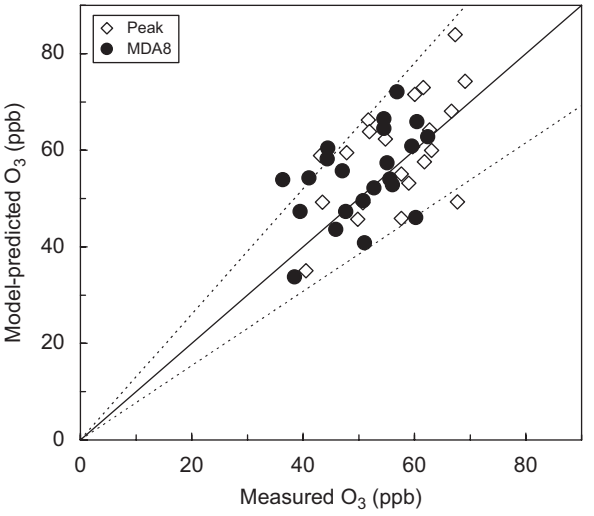

Fig. 5. Model-predicted average daily peak and MDA8 July O_3 versus measured values for all 22 locations. Lines are 1:1 ± 30%.

Table 5
Average measured and predicted concentrations, biases, and errors for ozone metrics

Ozone metric	Measured	Modeled	Bias	Error	Fractional bias	Fractional error
MDA8 [O_3]	50.7 ppb	54.5 ppb	+3.9 ppb	7.4 ppb	+0.07	0.14
Daily peak [O_3]	56.7 ppb	59.4 ppb	+2.7 ppb	7.9 ppb	+0.05	0.14
Hours >80 ppb	11.0 h	20.2 h	+9.2 h	14.5 h	+0.59	0.93
Hours >70 ppb	31.4 h	38.7 h	+7.3 h	21.9 h	+0.21	0.62

well by the model. The number of hours over 70 or 80 ppb, however, was predicted with considerably less accuracy.

4.3. $PM_{2.5}$

The model's ability to capture concentrations of total $PM_{2.5}$, sulfate, nitrate, ammonium, and organics was analyzed separately. Mean concentrations, variances of the set of monthly averages, and the distributions of 24-h average concentrations were examined.

4.3.1. Total $PM_{2.5}$

The distributions of daily average $PM_{2.5}$ concentrations for the simulated Januaries and Julys and for three measured Januaries and Julys (the three years during which both STN and IMPROVE measurements were made, 2002–2004) are shown in Fig. 6. The January distributions show less variability between years and closer agreement with measured distributions compared to the July distributions, especially for the upper two-thirds of daily concentrations. The biases and errors for model-predicted $PM_{2.5}$ concentrations are shown in Table 7. Predicted January concentrations generally showed a small positive bias, while there was a larger, generally negative, bias in July. This discrepancy between seasons is due in large part to the underprediction of organics and, to a lesser extent, sulfate in July. The biases and errors are similar to those calculated by Karydis et al. (2007) for simulation in PMCAMx of July 2001 and smaller than those calculated for simulation of January 2002. The July biases and errors are also somewhat less than those seen by Gaydos et al. (2007) for July 2001.

A comparison between model-predicted and measured average January and July $PM_{2.5}$ concentrations for the 22 locations is shown in Fig. 7. The large majority of model-predicted average concentrations were within 30% of measured values. This figure also shows the extent of the positive bias in January and the negative bias in July. January concentrations tended to show overpredictions in rural areas in January, while in July underpredictions occurred at most locations. January concentrations were overpredicted in 12 locations and underpredicted in 10, while July concentrations were overpredicted in four locations and underpredicted in 18. The model also captured the interannual variability of $PM_{2.5}$ concentrations reasonably well. There was no significant difference (F-test with $p<0.05$) between the variance in GRE-CAPS predicted concentrations and the variance in measured concentrations in 19 locations in January and 18 locations in July.

The quality of $PM_{2.5}$ predictions is summarized in Table 6. Model performance was "excellent" in January and "good" in July. In their PMCAMx simulations of January 2002 and July 2001, Karydis et al. (2007) had an "average" performance for January $PM_{2.5}$ and "good" performance for July $PM_{2.5}$. Therefore, GRE-CAPS performs at least as well as does standard retrospective modeling in simulating January and July total $PM_{2.5}$ concentrations.

Table 6
Overall GRE-CAPS performance

O_3	Peak O_3	Excellent[a]
	MDA8 O_3	Excellent
	Hours >70 ppb	Average
	Hours >80 ppb	Problematic
$PM_{2.5}$	Sulfate	
	January	Good
	July	Good
	Nitrate	
	January	Good
	July	Good
	Ammonium	
	January	Excellent
	July	Excellent
	STN organics	
	January	Good
	July	Average
	IMPROVE organics	
	January	Good
	July	Excellent
	Total $PM_{2.5}$	
	January	Excellent
	July	Good

[a]Excellent: |fractional bias| ⩽15% and fractional error ⩽35%; Good: |fractional bias| ⩽30% and fractional error ⩽50%; Average: |fractional bias| ⩽60% and fractional error ⩽75%; Problematic: |fractional bias| >60% or fractional error >75%.

4.3.2. Sulfate

The concentrations of sulfate tended to be overpredicted in January and underpredicted in July. The positive January bias and negative July bias are listed in Table 7. The fractional bias was <25% in both months, however, indicating rather unbiased model predictions. Predictions in most locations fall within 30% of measured values.

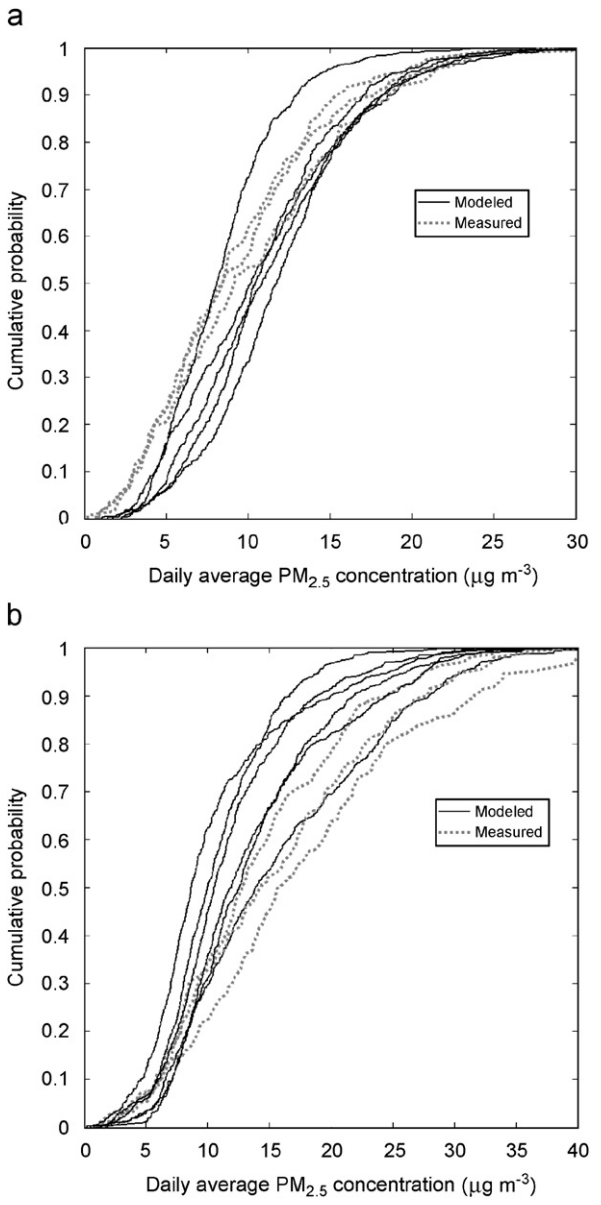

Fig. 6. Cumulative distribution functions of daily average $PM_{2.5}$ concentrations for (a) three measured and five model-predicted Januaries, and (b) three measured and six model-predicted Julys.

The biases for sulfate predictions in the GRE-CAPS model are close to those seen by Gaydos et al. (2007) for July 2001 (approximately $-1.5\,\mu g\,m^{-3}$), though the mean error was smaller in GRE-CAPS. The biases in GRE-CAPS were very close to those calculated by Karydis et al. (2007) for both January (approximately $+0.4\,\mu g\,m^{-3}$) and July (approximately $-1.0\,\mu g\,m^{-3}$).

There were few significant discrepancies between the model-predicted and measured interannual variabilities of sulfate concentrations. The model significantly ($p<0.05$) overpredicted variances in just four locations in January; it significantly overpredicted variances in only three locations in July, and it significantly underpredicted variances in only three locations in July. These are a small fraction of the total number of locations, indicating that GRE-CAPS performed well in simulating the interannual variability of sulfate.

Using the criteria outlined in Table 6, the performance of GRE-CAPS in simulating sulfate concentrations was "good" in both January and July. Karydis et al. (2007) reported "good" model performance for sulfate for January 2002 and "average" performance for July 2001. These indicate that GRE-CAPS simulates average sulfate concentrations at least as well as does standard PMCAMx modeling.

4.3.3. Nitrate

GRE-CAPS-predicted nitrate concentrations had a small negative bias in both January and July (Table 7). Karydis et al. (2007) reported a negative bias with respect to STN measurements and a positive bias with respect to IMPROVE measurements for a PMCAMx simulation of January 2002 as well as a negative bias for both networks in July 2001. Since the measurements in this study are dominated by STN sites, the negative bias is consistent with the findings of Karydis et al. (2007). The fractional errors of the GRE-CAPS model, however, were smaller than those of retrospective PMCAMx modeling (Karydis et al., 2007). Standard PMCAMx fractional errors were roughly 0.7 in January 2002 and 0.9 in July 2001, compared to 0.43 in January and 0.35 in July for GRE-CAPS.

GRE-CAPS tended to underpredict nitrate at urban sites and overpredict nitrate at rural sites in January. Karydis et al. (2007) noticed similar trends in their simulation of January 2002. These differences were attributed to difficulties with heterogeneous nighttime formation of nitrate in urban areas and overestimation of ammonia emissions in rural areas (Karydis et al., 2007). Nitrate was overpredicted at nine sites in January, compared to 13 underpredictions. In July, there were 11 of each. In January, there was no significant difference between the GRE-CAPS predicted interannual variability and the measured interannual variability in 16 locations. There were significant differences between modeled and measured nitrate variabilities in half of the locations in July, but this is largely due

Table 7
Average measured and predicted concentrations, biases, and errors

Species	Month	Measured ($\mu g\,m^{-3}$)	Predicted ($\mu g\,m^{-3}$)	Mean bias ($\mu g\,m^{-3}$)	Mean error ($\mu g\,m^{-3}$)	Fractional bias	Fractional error
Sulfate	January	2.43	2.86	+0.42	0.53	+0.16	0.20
	July	5.37	4.25	−1.13	1.28	−0.23	0.27
Nitrate	January	1.94	1.56	−0.38	0.75	−0.21	0.43
	July	0.49	0.42	−0.07	0.16	−0.16	0.35
Ammonium	January	1.31	1.38	+0.07	0.26	+0.05	0.20
	July	1.62	1.59	−0.03	0.30	−0.02	0.19
Organics	January	4.25	3.13	−1.12	1.64	−0.30	0.44
(STN)	July	5.31	3.09	−2.23	2.23	−0.53	0.53
Organics	January	1.70	2.12	+0.42	0.82	+0.22	0.43
(IMPROVE)	July	2.53	2.73	+0.20	0.30	+0.08	0.12
Total $PM_{2.5}$	January	10.46	10.80	+0.34	2.12	+0.03	0.20
	July	15.68	12.71	−2.97	3.44	−0.21	0.24

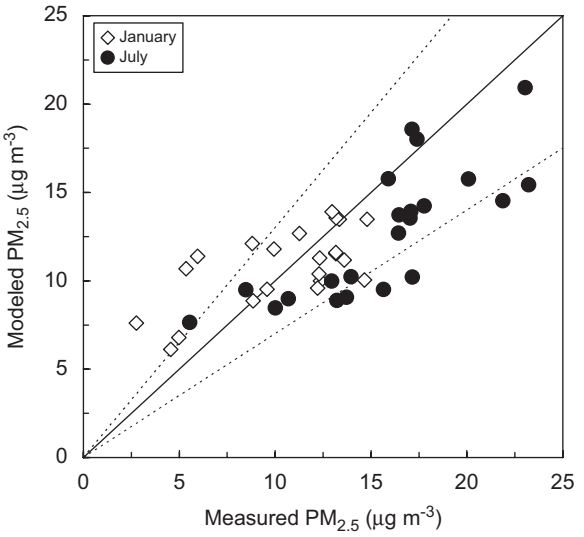

Fig. 7. Model-predicted average January and July $PM_{2.5}$ concentrations versus measured average January and July $PM_{2.5}$ for all 22 locations. Lines are 1:1 ± 30%.

to the very small July nitrate concentrations in the Eastern USA.

The ability of GRE-CAPS to simulate nitrate concentrations is described as "good" using the criteria in Table 6 for both January and July. Karydis et al. (2007) had "average" performance for January 2002 and "problematic" performance for July 2001. GRE-CAPS predictions had smaller fractional errors than did standard PMCAMx simulations, resulting in the difference in performance classifications. GRE-CAPS simulations, therefore, describe January and July nitrate concentrations at least as well as standard retrospective modeling in PMCAMx.

4.3.4. Ammonium

Model-predicted ammonium concentrations were the closest to measurements of all the PM species (Table 7). This accuracy is likely due in large part to the ammonia emissions inventory of Pinder et al. (2004), which was used in the model. This ability to predict ammonium emissions accurately was also seen by Karydis et al. (2007), who used the same ammonia inventory. For the 19 sites at which ammonium was measured, there were 12 overpredictions and 7 underpredictions in January and 10 overpredictions and 9 underpredictions in July. There was no evident pattern as to which locations had underpredictions and which had overpredictions. The Shenandoah site had relatively large overpredictions in both months ($0.5\,\mu g\,m^{-3}$ in January and $1.0\,\mu g\,m^{-3}$ in July), while the Kansas City site had rather large underpredictions in both months ($-0.5\,\mu g\,m^{-3}$ in January and $-0.6\,\mu g\,m^{-3}$ in July).

The interannual variability of ammonium concentrations was also captured well by GRE-CAPS. There were significant overpredictions of variances of monthly average concentrations in just three locations in January, while in July there was one significant overprediction and one significant underprediction. These indicated that there is little difference between the interannual variabilities of measured and model-predicted ammonium concentrations.

The small errors and biases in ammonium predictions led to a classification of "excellent" for

GRE-CAPS model performance for ammonium (Table 6). The mean and fractional biases and errors led to better performance by GRE-CAPS than for stand-alone PMCAMx. Model performance according to Karydis et al. (2007) for ammonium predictions was "good" for January 2002 and "average" for July 2001.

4.3.5. Organic PM

There were large discrepancies in biases and errors in organic PM concentrations between January and July, and between STN and IMPROVE sites (Table 7). The mean biases and errors were larger at the STN sites than at the IMPROVE sites. Karydis et al. (2007) calculated positive biases for PMCAMx-predicted organics at both STN and IMPROVE sites in January 2002 and negative biases for both networks in July 2001. Lane et al. (2006) suggested that the overprediction in January OM concentrations could be due to an overestimation of emissions due to wood burning. Karydis et al. (2007) attribute part of this discrepancy to the lack of correction for sampling artifacts in the STN measurements. Very close agreement between the model and measurements would be somewhat surprising, considering the lack of any volatility of primary OM in the model (Robinson et al., 2007), the crude treatment of biogenic SOA precursors (with only a single gas-phase biogenic precursor), and the lack of oligomerization reactions in the condensed phase in the model (Karydis et al., 2007).

Concentrations of organic PM were underestimated at 10 of 16 STN locations in January and at all 16 STN locations in July. In contrast, organic concentrations were overestimated at 5 of 6 IMPROVE locations in both months. The large discrepancies in model performance with respect to the two networks among even rural sites indicate that there is likely an effect of the collection method and the uncertainty in measurements in the model biases and errors. The crude method of converting OC to OM by simply multiplying by 1.4 can also introduce errors.

In spite of the somewhat large biases and errors in predicted OM concentrations, GRE-CAPS was able to simulate the interannual variability of OM concentrations rather well. In January, the model-predicted variance was significantly greater than measured variances in 1 of the 22 locations and significantly less than measured variances in 3 locations. In July, when errors and biases in OM predictions were largest, the variance did not differ significantly from the measured variance in 18 locations.

The quality of OM predictions by GRE-CAPS spanned the range from "average" to "excellent" (Table 6). Performance was best at IMPROVE sites, possibly due to their rural locations and possibly due in part to the collection method and artifact correction at IMPROVE sites. These predictions are of similar quality to those of Karydis et al. (2007), in which July predictions were considered "good" and January predictions were considered "average".

5. Conclusion

The GRE-CAPS modeling system was used to model present-day concentrations of O_3 and $PM_{2.5}$ over the Eastern USA. The modeling system used a global GCM/CTM (GISS II′) to generate meteorology and simulate intercontinental chemical transport. Meteorology was downscaled using a regional meteorological model (MM5), and air quality was simulated using a regional CTM (PMCAMx). Five present-day Januaries and six present-day Julys were simulated. Predicted O_3 concentrations were compared to 5 years of measured data (2001–2005), while predicted $PM_{2.5}$ concentrations were compared to 4 years of measured data (2001–2004 for IMPROVE sites, 2002–2005 for STN sites).

GRE-CAPS predictions of O_3 concentrations compared well to measured concentrations. Model performance for GRE-CAPS was of similar quality to that of PMCAMx when used for standard retrospective episode modeling. GRE-CAPS was able to capture accurately longer-term climatological average concentrations, which have not previously been investigated using standard PMCAMx. Model-predicted daily peak and MDA8 O_3 concentrations had small biases and were close to measured values, resulting in "excellent" model performance. Daily peak O_3 had a mean bias of $+2.7$ ppb, while MDA8 O_3 had a mean bias of $+3.9$ ppb. The O_3 metrics used to represent O_3 episodes, hours with O_3 concentrations >70 or 80 ppb, did not agree as well with measurements. The model also had a tendency to overpredict the interannual variability of O_3 concentrations, mostly as the result of a single July with very high predicted O_3 levels in the Southeastern USA. The distributions of model-predicted daily peak and MDA8 O_3 concentrations were similar to the distributions of the measured concentrations and agreed with the

distribution of measurements approximately as well as the modeling system of Hogrefe et al. (2004a, b).

The model predicted concentrations of the various PM species with reasonable accuracy. The biases and errors in model-predicted sulfate, nitrate, ammonium, and organics are generally similar to those of Gaydos et al. (2007) and Karydis et al. (2007) for their PMCAMx simulations of July 2001 and January 2002. Additionally, the biases in GRE-CAPS predictions of O_3 and $PM_{2.5}$ concentrations were somewhat smaller than those of the modeling system of Tagaris et al. (2007). The categorical descriptions of the quality of the model were similar to those of Karydis et al. (2007).

The performance of the GRE-CAPS modeling system is promising, with biases and errors in monthly average concentrations similar to those calculated when simulating actual historical time periods. The results suggest that the GRE-CAPS system adequately captures the mean and inter-annual variability of O_3 and $PM_{2.5}$ concentrations. Future work will focus on using this system with predictions of future climate, intercontinental transport, and emissions to estimate how changes in these factors will affect air quality in the Eastern USA.

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